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14. ABSTRACT

Transparent armor that mitigates blast and high-speed projectile is critical for personnel protection, and can be used to reinforce windows in buildings, ground/air vehicles, and military installations. A multilayer architecture, such as those found in naturally occurring nacre shell, can greatly reduce weight and enhance the mechanical strength and toughness. However, optical transmission of layered materials degrades dramatically as the number of layers increases, limiting visibility. Light reflections in multilayers also causes thin-film interference, resulting in the

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Enhancing Optical Transmission of Multilayer Composites with Interfacial Nanostructures

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1. Statement of the Problem Studied

Transparent armor that mitigates blast and high-speed projectile is critical for personnel protection, and can be used to reinforce windows in buildings, ground/air vehicles, and military installations. A multilayer architecture, such as those found in naturally occurring nacre shell [1-8], can greatly reduce weight and enhance the mechanical strength and toughness. However, optical transmission of layered materials degrades dramatically as the number of layers increases, limiting visibility. Light reflections in multilayers also causes thin-film interference, resulting in the iridescent appearance commonly observed nacre shells. The objective of the proposed research is to investigate the fabrication and optical properties of a new class of multilayer ceramic-polymer composites with interfacial nanostructures. Fresenl reflection is a common physical phenomenon caused by the discrete mismatch in refractive index at a material interface. Inspired by the moth eye [9-15], this reflection can bes suppressed by introducing a finite medium with gradient index that gradually bridge the index mismatch. The proposed research introduces nanostructures at the interface of two dissimilar materials to enhance optical transmission (**Figure 1**). The design and experimental tools developed in this research will further the understanding of nanoscale light interactions and enable multilayer ceramic-polymer composite armor with broadband and wide-angle optical clarity. This work can contribute towards the Army's mission in blast/projectile mitigation, and lead to lightweight transparent armor with enhanced toughness and broadband optical transmission.

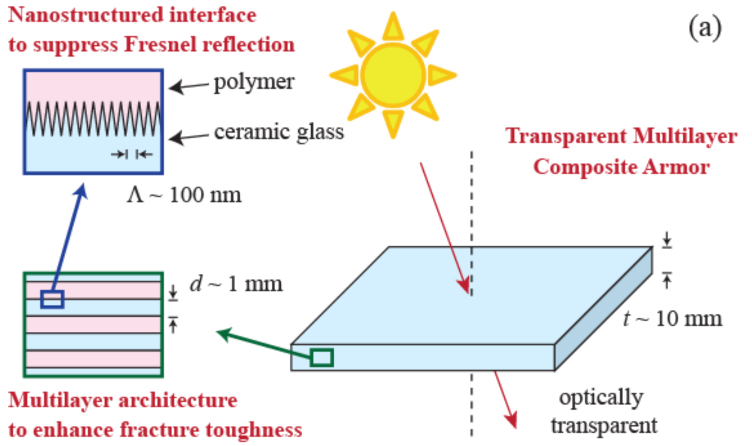

Figure 1. The proposed multilayer transparent composite armor with enhanced optical transmission.

2. Summary of the Most Important Results

In this project we successfully 1) constructed rigorous optical models, 2) developed fabrication and assembly processes to created nanostructured multilayers, and 3) demonstrated experimentally that light reflections in a silia-polymer interface can be suppressed using embedded nanostructures [16]. The fabricated prototype exhibits enhanced transmission for broad wavelength (400-800 nm) and wide angle (0-75º), and also shows suppressed iridescent effects. A representative top-view scanning electron microscope (SEM) image of a fabricated double-side patterned nanocone structure in silica substrate is shown in **Figure 2**. The structure was patterned using a custom-built interference lithography (IL), where nanostructures with ~100 nm feature size, 220 nm period, and 400 nm tall were patterned and etched onto both sides of a ~1 mm-thick silica substrate. The reactive-ion etching (RIE) process utilizes CHF$_3$, which increased the structure aspect ratio as it was etched into silica. We have also explored other RIE chemistries using HBr and CF$_4$ using poly-crystalline silicon as an intermediate mask with limited success. The resulting structure height is sufficient for AR properties in the visible, but would need to be taller for operation in the infrared.

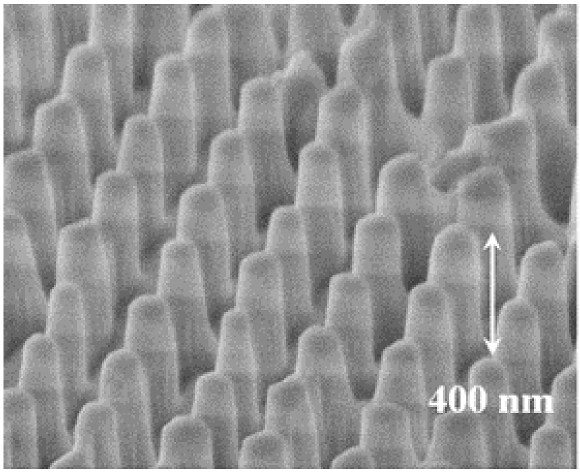

Figure 2. Fabricated double-side silica surface nanostructures [16].

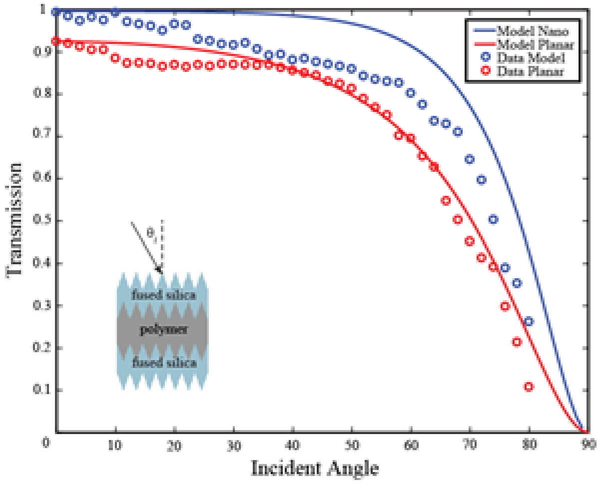

Figure 3. Measured angle-dependent transmission of silica/polymer/silica stack [16].

A stacking and bonding process was developed to assemble and bond individual double-side patterned silica substrates into multilayer composites. In this process two double-side patterned silica substrates were bonded by a UV-curable polymer from Norland optical adhesive (NOA). Edge spacers are used to control the thickness and uniformity of polymer layers. This generates a silica-polymer-silica stack, which can be bonded repeatedly to yield a thicker multilayer panel. Using the developed stacking and bonding process, the double-side patterned sapphire substrates will be assembled into multilayer element. This parallel assembly process allows precise control of layer thickness and defect inspection between layers, and can be scaled to large prototypes for mechanical testing. In this research the process was used to fabricate a silica-polymer-silica prototype with nanostructures at all interfaces and surfaces.

The fabricated silica-polymer-silica composite demonstrates enhanced optical properties, as shown in **Figure 3** Here the specular transmission of a 633 nm wavelength, TE-polarized laser was measured vs incident angles for fabricated samples with planar and nanostructured interface. **Enhancement from 10-15% can be observed** at various angles, illustrating successful suppression of Fresnel reflection. The experimental data agrees well to the constructed multilayer optical model based on rigorous coupled-wave analysis (RCWA). The lower transmission can be attributed to non-ideal structure profile and fabrication defects, which can be improved by optimizing the etching processes. We have also examined the theoretical enhancement as the number of layers are further increased. Defining a single silica-polymer-silica stack as one period (N=1), the optical transmission for N=10 and 20 is depicted in **Figure 4**. Here it can be observed that while transmission drops significantly for the multilayer with planar interfaces,

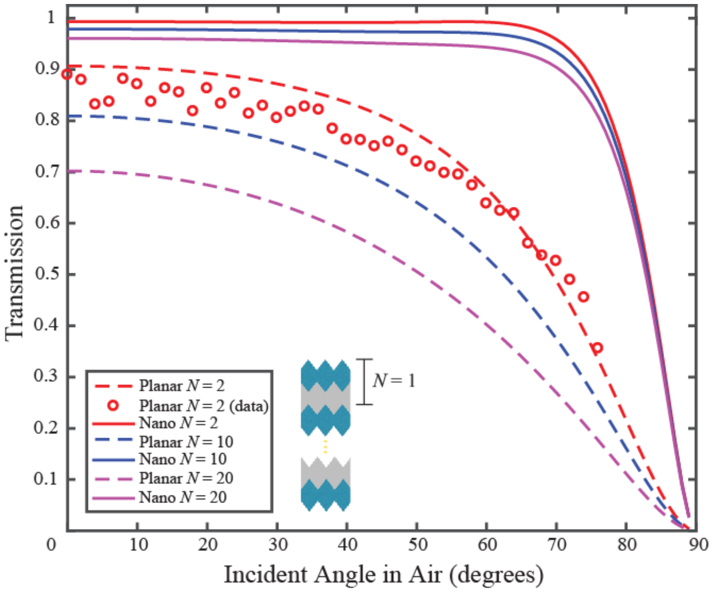

Figure 4. Theoretical transmission for N composite layers.

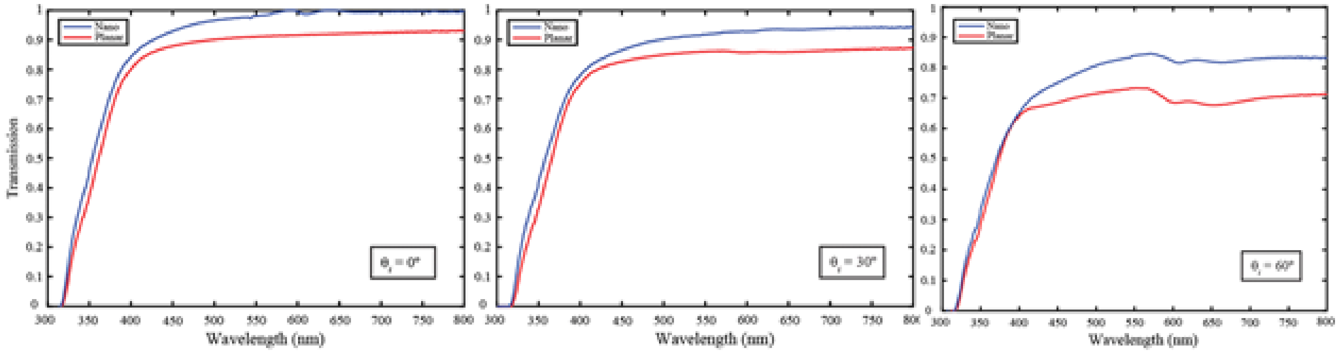

Figure 5. Experimentally measured transmittance for incident angles of 0, 30, and 60 degrees [16].

those with nanostructures will yield low degradation in optical transmission. This is especially evident in the high angle range, where transmission can be maintained at 90% at ~75%, as oppose to 20% for the planar N = 20 case. Future work includes experimental data on high N layers to validate the theoretical prediction and demonstrate layer scalability.

The transmittance of the fabricated silica-polymer-silica sample from UV to near-IR was also characterized to investigate the broadband response. The transmittance at incident angles of 0, 30, and 60 degrees for the composite stack with nanostructured and planar (reference) interfaces are shown in **Figure 5**. It can be observed that the structure exhibits broadband transmission enhancement from 400–800 nm. The enhancement varies from 10 to 15% for normal and larger incident angles, respectively, consistent with the laser measurement shown in **Figure 3**. This demonstrate that the visibility through the multilayer composite would maintain broadband clarity even at various viewing angles.

We also theoretically and experimentally examined the suppression of iridescence effects. In this system we examined the interference of reflected light in a thin film that is a few wavelength thick so the model would be valid for low-coherence light sources, such as ambient sunlight and artificial lighting. The broadband transmission of a glass and polymer thin films, each 500 nm thick, on a silica substrate with planar and nanostructured interfaces are simulated in **Figure 6**. Here the nanostructured sample contains a linear taper nanostructure with 500 nm height. It can be observed that intensity oscillation is significant in the planar sample case, indicating

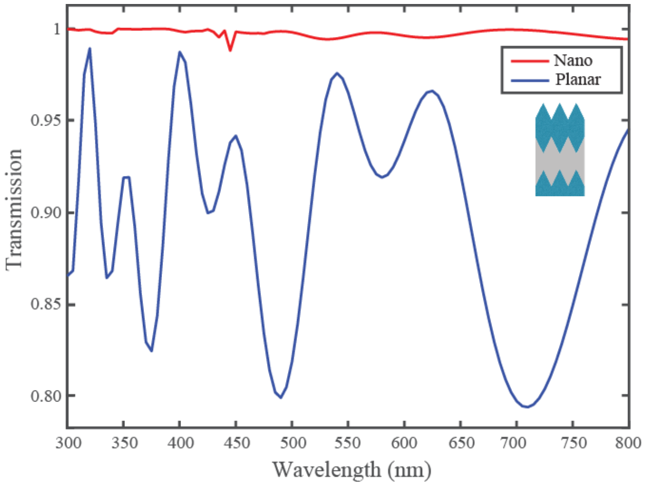

Figure 6. Fabricated double-side silica surface nanostructures [16].

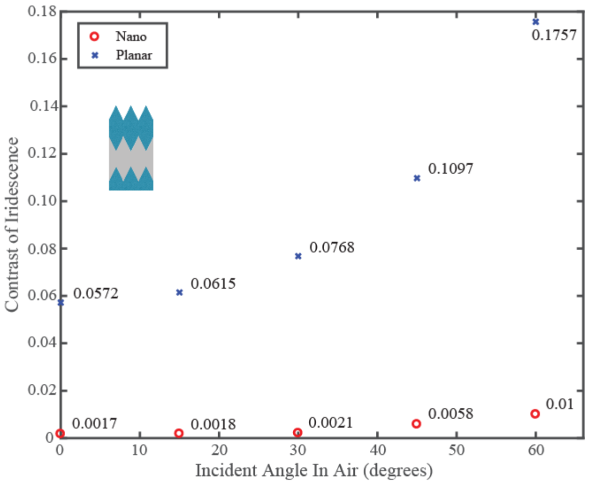

Figure 7. Measured angle-dependent transmission of silica/polymer/silica stack [16].

wavelength-selective behavior that results in appearance of color. The nanostructured sample, on the other hand, have a near-constant response, resulting in suppression of iridescence. **Figure 7** illustrates the fringe contrast, a measure of interference by looking at the ratio of AC vs DC components, for both samples. It can be observed that contrast can be suppressed by 1-2 order-of-magnitude, with bigger improvement at higher incident angles. It is also interesting to note that for the nanostructured sample the iridescence is fairly constant, while for planar composites it grows more wavelength selective at higher viewing angles.

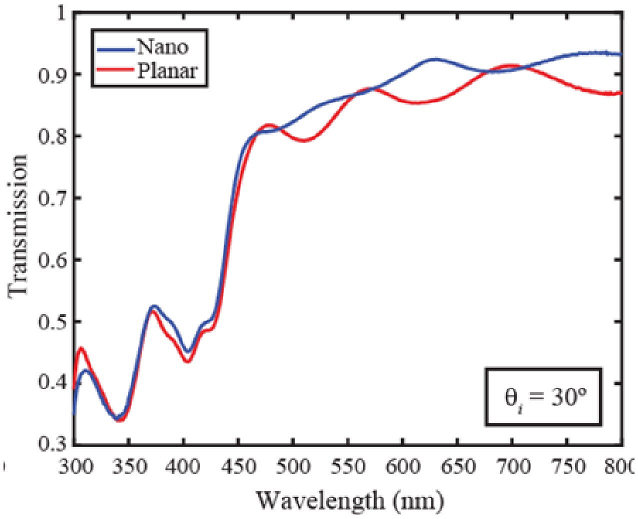

Figure 8. Reflectance of thin-film sample with planar vs nanostructured interface [16].

The suppression of sample iridescence was examined experimentally by fabricating a single-side patterned silica substrate, which is then coated with a thin polymer layer approximately 700 nm thick. The broadband transmittance of the thin-film samples with nanostructured and planar interfaces are shown in **Figure 8**. It can be observed that the transmittance for the nanostructured interface sample has lower intensity oscillation, which is characteristic of thin-film interference. The nanostructured sample's fringe contrast was quantitatively measured to be suppressed by a factor of three. While this is smaller than predicted, it demonstrates significant suppression of iridescence effects. Using constructed optical model, we predicted that the interference effects can be suppressed further by using taller interfacial nanostructures with a more graduate taper.

In this work we demonstrate the basic principle that interfacial nanostructures in multilayer composites can reduce Fresnel reflection. The fabricated silica-polymer-silica prototype exhibits 10-15% transmission enhancement across 400-800 nm wavelength and incident angles of 0-75°, and a three-fold reduction in interference contrast. This work can lead to multifunctional composites with enhanced mechanical strength, toughness, and optical clarity, and can potentially revolutionize DoD's capability in anti-blast/projectile transparent armor. Future work will focus on ceramic materials more suitable for armor applications (sapphire, AlON, etc.) increasing structure height, and assembling $N > 5$ layers to demonstrate systematic improvement.

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